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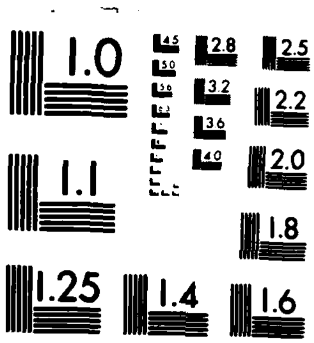

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Annual summary report is presented on experimental investigations of molecular interactions on model catalyst surfaces using high-resolution electron energy loss spectroscopy (EELS). Data on acetylene and benzene adsorption on palladium surfaces is described.

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OFFICE OF NAVAL RESEARCH

Contract N00014-80-C-0147

Annual Report No. ~~4~~61

ANNUAL SUMMARY REPORT - 1984 - CONTRACT NO. N00014-80-C-0147

by

L.L. Kesmodel

10 October 1984

Report: Annual Summary

Contract No.: N00014-80-C-0147

Title: Vibrational Spectroscopy of Chemisorption Phenomena on Model Catalyst Surfaces

Principal Investigator: Dr. Larry L. Kesmodel, Indiana University

Project Description

The technique of high-resolution electron energy loss spectroscopy (EELS) is used to define the nature of hydrocarbon adsorption and reactions on model catalyst surfaces in controlled ultrahigh vacuum environments. The EELS method provides detailed vibrational spectra of adsorbed species on surfaces in the 100-4000 cm^{-1} energy loss range. Supporting information is obtained from Auger spectroscopy, thermal desorption spectroscopy (TDS) and low-energy electron diffraction (LEED).

The goal of this project is to increase our understanding of the fundamental behavior of hydrocarbon bonding and chemical reaction behavior on metal surfaces acting as model catalysts.

Scientific Progress

Research this past contract year has centered on the interaction of the prototype hydrocarbons acetylene (C_2H_2) and benzene (C_6H_6) with single crystal (111) and (100) surfaces of palladium metal as outlined below.

(1) Thermal Evolution and Decomposition of Acetylene

We have obtained extensive EELS data on the high temperature behavior (300-500K) of acetylene on both Pd(100) and Pd(111) surfaces.(1) Of particular interest is the formation of CCH species on both surfaces, which we identified spectroscopically for the first time in this context. On Pd(100), CCH forms with thermal processing of chemisorbed acetylene to 400K but on Pd(111) is co-adsorbed with a $\equiv C-CH_3$ (ethylidyne) species following

300K C_2H_2 adsorption. With further annealing to 450K, CCH is the dominant species on both surfaces. This evidence for CCH formation may impact on earlier studies on Rh, Pt, and Ni where CH formation was instead postulated. Our work suggests that dehydrogenation is the preferred decomposition path of acetylene and that CC bond scission may not occur until higher temperatures.

We are also currently studying the possible formation of benzene (C_6H_6) from acetylene on Pd(111). This reaction was reported by Tysoe _et al._[2] but we have not as yet been able to confirm this result using thermal desorption spectroscopy. To complete this study we are upgrading our sample dosing system with a multichannel array doser.

(2) Benzene Chemisorption on Pd(100 and Pd(111)

We have investigated the adsorption of C_6H_6 and C_6D_6 on the (100) and (111) surfaces of palladium using EELS and TDS.[3,4] The EELS spectra indicate that the benzene molecule is bonded with the ring parallel to the surface, and that the molecule is not strongly perturbed from the gas phase. Thermal desorption spectra indicate a competing process of benzene desorption and decomposition, with decomposition favored at low initial coverages ($\lesssim$ 0.5 Langmuir).[4]

We have also studied the mechanism of electron scattering for chemisorbed benzene and have recently reported evidence for a resonance scattering mechanism.[3] We find evidence for mode-selective vibrational excitation as a function of electron beam energy which is characteristic of negative-ion resonance scattering. Our observations constitute the most extensive and compelling evidence to date for resonance scattering in a chemisorption system. Resonance scattering may be very important in EELS

studies of adsorption due to the large vibrational cross sections and the ability to observe overtone excitations.

(3) Equipment Development

A unique facility for high-resolution EELS studies of vibrational spectra has been developed at Indiana University for use in the research project. This facility includes two ultrahigh vacuum systems with base pressures in the 10^{-11}-10^{-10} Torr range, each with LEED, Auger, ion-sputtering, and EELS capability and one system with a computer-controlled quadrupole mass spectrometer for desorption studies.

Each of the EELS instruments is of the 127° cylindrical deflection design and routinely achieve resolution in the 5-8 meV range. However, one of the instruments is of a new design(5) and has achieved resolution as low as 2.5 meV. This instrument also exhibits much higher signal levels at a given energy resolution than other competing EELS instrumentation.(5)

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